 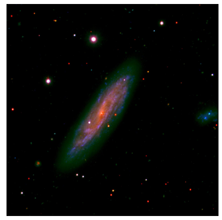 LBT & Image

ATOMIC SPECTROSCOPY: ASTRONOMY TO BIO-MEDICAL SCIENCE

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"63rd International Symposium on
Molecular Spectroscopy"
Ohio State University, Columbus, Ohio, USA
June 16-20, 2008

SPECTROSCOPY IN ASTRONOMY AND MEDICAL SCIENCE

- **ASTRONOMY:**

- Astronomical objects are studied in two ways:

  - **Photometry:** - Beautiful pictures or images of astronomical objects, Stars, Nebulae, Active Galactic Nuclei (AGN), Blackhole Environments, etc
  - Bands of Electromagnetic Colors ranging from X-ray to Radio waves → macroscopic information

  - **Spectroscopy:** - Provides most of the detailed knowledge: temperature, density, extent, chemical composition, etc. of astronomical objects

- **NANOSCIENCE:**

  - X-ray Spectroscopy in Cancer Research

Spectroscopy is underpinned by Atomic & Molecular Physics

BLACKHOLE JET OF CENTAURUS A

(Observed by Chandra space telescope)

• Photometric image: red - low-energy X-rays, green - intermediate-energy X-rays, and blue - the highest-energy X-rays. The dark green and blue bands are dust lanes that absorb X-rays.

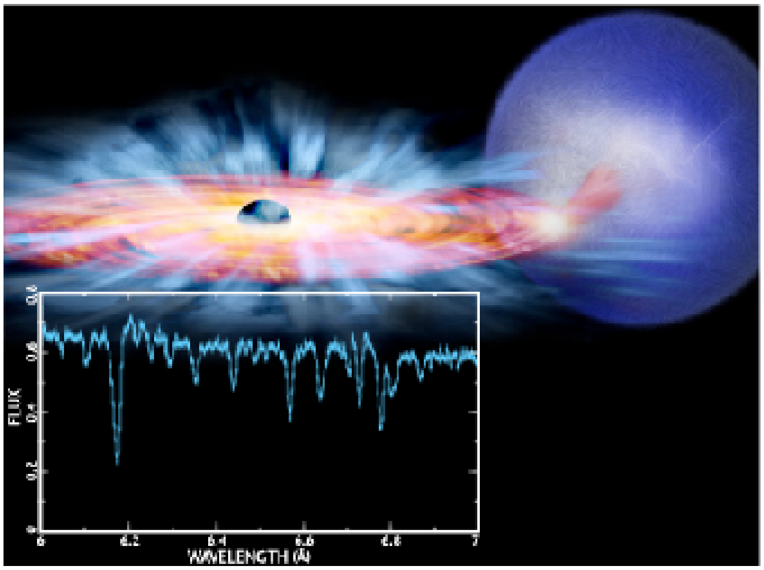
• Materials from nearby stars sucked into the blackhole & ejected as a jet (L & E conservation)

• Blasting from the black hole in the galaxy a jet of a billion solar-masses extending to 13,000 light years

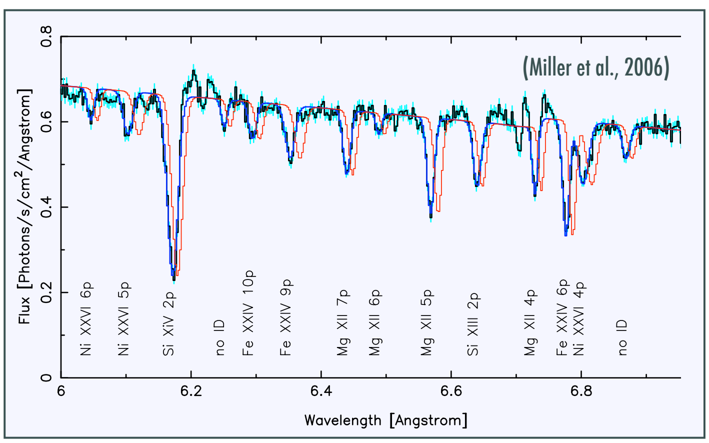
• The falling particles spiral around the blackhole, move faster close to it and release energy in the form of radiation

• The highly energetic atoms - SUPERHOT ATOMS - near the blackhole are in a plasma state & emit bright X-rays

SPECTRUM of the Wind near Blackhole: GRO J1655-40 Binary Star System



- **Materials from the large star is sucked into the blackhole** - form wind as they spiral to it



Spectrum: Highly charged Mg, Si, Fe, Ni lines
Red Spectrum - Elements in natural widths
Doppler Blue Shift - Wind is blowing toward us

DOMINANT RADIATIVE ATOMIC PROCESSES IN ASTROPHYSICAL PLASMAS

and Relevant Atomic Parameters

1. Photoexcitation & De-excitation (bound-bound transition):

$$X^{+Z} + h\nu \rightleftharpoons X^{+Z*}$$

- **Oscillator Strength (f), Radiative Decay Rate (A-value)**
- **Examples:** **Seen as lines in astrophysical spectra**
- **Determines opacities in astrophysical plasmas**

2. Photoionization (PI) & Radiative Recombination (RR):

$$X^{+Z} + h\nu \rightleftharpoons X^{+Z+1} + \ e$$

3. Autoionization (AI) & Dielectronic recombination (DR):

$$e + X^{+Z} \rightleftharpoons (X^{+Z-1})^{**} \rightleftharpoons \begin{cases} e + X^{+Z} & \mathrm{AI} \\ X^{+Z-1} + h\nu & \mathrm{DR} \end{cases}$$

The doubly excited state - "autoionizing state" - introduces resonances

- 2 & 3. **Photoionization Cross Sections (σ_{PI}), Recombination Cross Sections (σ_{RC}) and Rate Coefficients (α_{RC})**

Examples:

- **Photoionization resonances - seen in absorption spectra,**
- **Recombination resonances - seen in emission spectra**
- **Determine ionization fractions in astrophysical plasmas**

Plasma Opacities - Radiation Transport

Monochromatic opacity (κ_ν) depends primarily on:

i) Bound - bound transitions (oscillator strengths, f_{ij})

$$\kappa_\nu(\mathbf{i} \rightarrow \mathbf{j}) = \frac{\pi \mathbf{e^2}}{\mathbf{mc}} \mathbf{N_i f_{ij}} \phi_\nu$$

N_i **= ion density in state i,** ϕ_ν **is a profile factor**

ii) Bound - free transitions (photoionization cross sections, σ_{PI})

$$\kappa_\nu = \mathbf{N_i} \sigma_{\mathbf{PI}}(\nu)$$

- **The opacity depends on interaction of radiation with all atoms and in all ionization stages.**

- **About 25 years ago, it was realized the existing opacities, from atomic data using mainly simple approximations, were incorrect by factors of 2 to 5 resulting in inaccurate stellar models. For example, Cepheid stars, which are important to determine distances of astronomical objects, could not be modeled.**

A plea was made for accurate opacity from accorate atomic parameters.

THE OPACITY PROJECT & THE IRON PROJECT:
AIM: Accurate Study of Atomic Processes in Astrophysical Plasmas & Calculate Opacities

International Collaborations: France, Germany, U.K., U.S. (Ohio State U, NASA-Goddard, Rollins), Belgium, Venezuela, Canada

- **THE OPACITY PROJECT (OP) (1982 -): study radiative atomic processes and radiation transport in astrophysical plasmas - all elements from H to Fe**

- **THE IRON PROJECT - IP (1993 -): study collisional & radiative processes of Fe & Fe peak elements**

- **Atomic & Opacity Databases: TOPbase, TIPbase at CDS (France), Ohio Supercomouter Center (OSC)**
http://vizier.u-strasbg.fr/topbase/topbase.html,
http://opacities.osc.edu
NORAD - www.astronomy.ohio-state.edu/∼nahar/nahar_radiativeatomicdata/index.html

- **Results from the OP and the IP have solved and continue to solve many outstanding problems. For example, existence of blackholes. abundances of elements, opacities in astrophysical plasmas, missing mass calculations.**

THEORY: Close Coupling Approximation & R-matrix Method

For a multi-electron system, in nonrelativistic LS coupling:

$$H^{NR}\Psi = \left[\sum_{i=1}^{N}\left\{-\nabla_i^2 - \frac{2Z}{r_i} + \sum_{j>i}^{N}\frac{2}{r_{ij}}\right\}\right]\Psi = E\Psi. \qquad (1)$$

Relativistic effects: Breit-Pauli R-matrix (BPRM) approximation includes three one-body relativistic correction terms:

$$H_{N+1}^{\mathrm{BP}} = H_{N+1}^{NR} + H_{N+1}^{\mathrm{mass}} + H_{N+1}^{\mathrm{Dar}} + H_{N+1}^{\mathrm{so}}, \qquad (2)$$

$H^{mass} = -\frac{\alpha^2}{4}\sum_i p_i^4$, $H^{Dar} = \frac{\alpha^2}{4}\sum_i \nabla^2\left(\frac{Z}{r_i}\right)$, $H^{so} = \left[\frac{Ze^2\hbar^2}{2m^2c^2r^3}\right]\mathbf{L}.\mathbf{S}$

The spin-orbit interaction H^{so} splits LS energy in to fine structure levels.

For a multi-electron system, the two-body terms are introduced in the Breit-Pauli Hamiltonian:

$$H_{\mathrm{BP}} = H_{NR} + H_{\mathrm{mass}} + H_{\mathrm{Dar}} + H_{\mathrm{so}} +$$

$$\frac{1}{2}\sum_{i\neq j}^{N}[g_{ij}(so+so') + g_{ij}(ss') + g_{ij}(css') + g_{ij}(d) + g_{ij}(oo')]. \qquad (3)$$

where the Breit interaction is

$$H^B = \sum_{i>j}[g_{ij}(so+so') + g_{ij}(ss')] \qquad (4)$$

Wave functions and energies are obtained solving:

$$\mathbf{H\Psi = E\Psi}$$

- $\mathbf{E < 0} \rightarrow$ **Bound (e+ion) states** Ψ_B
- $\mathbf{E \geq 0} \rightarrow$ **Continuum states** Ψ_F

Close-coupling Approximation and the R-matrix method

- **In close coupling (CC) approximation, the ion is treated as a system of (N+1) electrons: a target or the ion core of N electrons with the additional interating (N+1)th electron:**
- **Total wavefunction expansion is expressed as:**

$$\Psi_E(e + ion) = A \sum_{i}^{N} \chi_i(ion)\theta_i + \sum_{j} c_j \Phi_j(e + ion)$$

$\chi_i \rightarrow$ **target ion or core wavefunction**
$\theta_i \rightarrow$ **interacting electron wavefunction (continuum or bound)**
$\Phi_j \rightarrow$ **correlation functions of (e+ion)**

- **The complex resonant structures in the atomic processes are included through channel couplings.**
- **Substitution of** $\Psi_E(e + ion)$ **in** $H\Psi_E = E\Psi_E$ **results in a set of coupled euqations**
- **Coupled equations are solved by R-matrix method**

ATOMIC PROCESSES: Quantity of Interest - S (Line Strength)

Transition Matrix elements:

$< \Psi_B||\mathbf{D}||\Psi_{B'} >$ $\rightarrow$ **Radiative Excitation and Deexcitation**
$< \Psi_B||\mathbf{D}||\Psi_F >$ $\rightarrow$ **Photoionization and Recombination**

$\mathbf{D} = \sum_i \mathbf{r}_i$ $\rightarrow$ Dipole Operator
The matrix element reduces to generalized line strength,

$$S = \left| \left\langle \Psi_f | \sum_{j=1}^{N+1} r_j | \Psi_i \right\rangle \right|^2 \tag{5}$$

PHOTO-EXCITATION AND DE-EXCITATION:
The oscillator strength (f_{ij}) and radiative decay rate (A_{ji}) for the bound-bound transition are

$$f_{ij} = \left[\frac{E_{ji}}{3g_i}\right] S, \quad A_{ji}(sec^{-1}) = \left[0.8032 \times 10^{10} \frac{E_{ji}^3}{3g_j}\right] S \tag{6}$$

PHOTOIONIZATION:
The photoionization cross section, σ_{PI},

$$\sigma_{PI} = \left[\frac{4\pi}{3c}\frac{1}{g_i}\right] \omega S, \tag{7}$$

ω $\rightarrow$ **incident photon energy in Rydberg units**

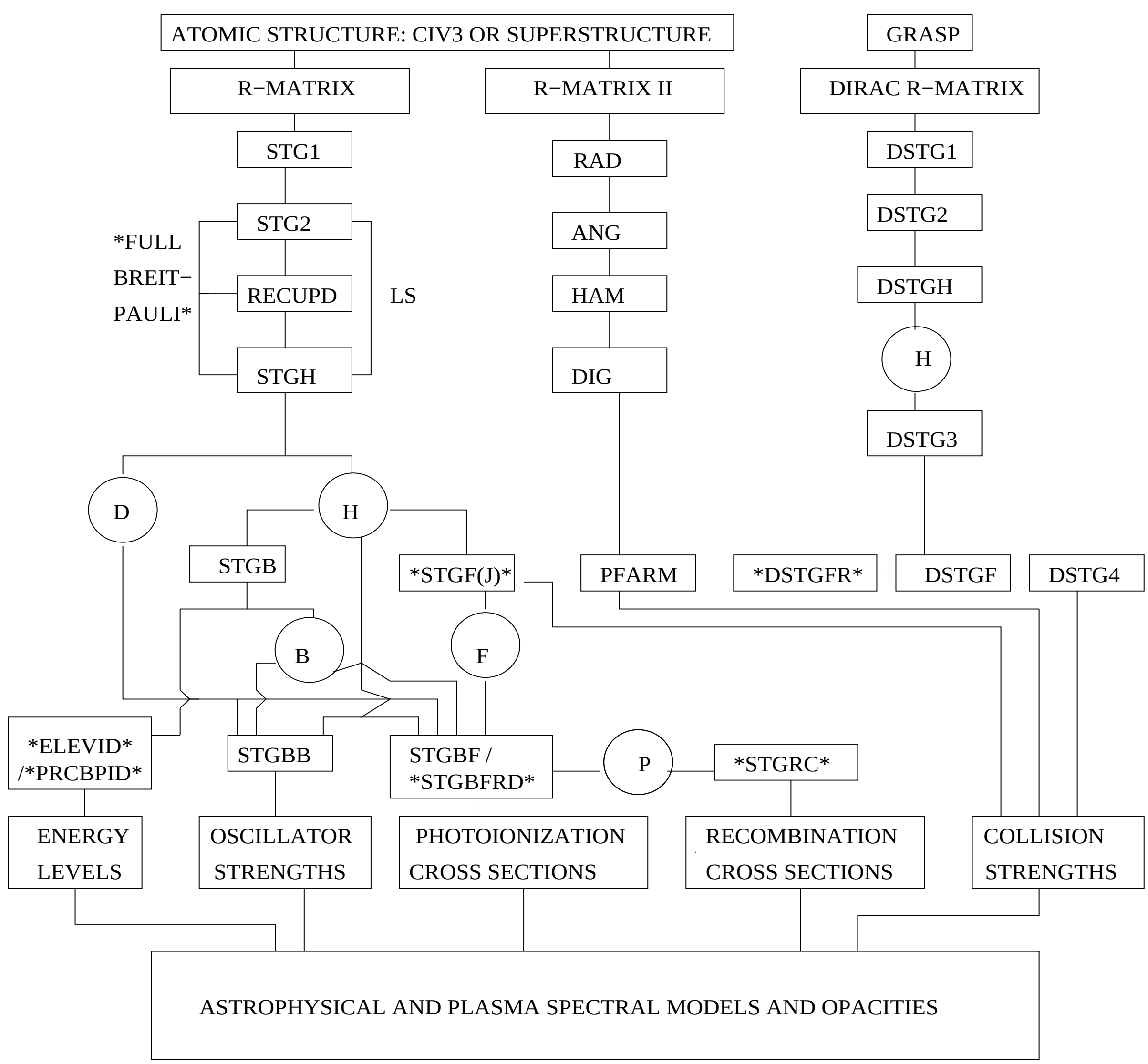
THEORETICAL SPECTROSCOPY OF TRANSITIONS:

- **Relativistic Briet-Pauli R-matrix calculations result in a large number of energy levels & transitions; however, without spectroscopic identification**
- **Theoretical spectroscopy for level identification is a major task. It is based on quantum defect analysis**

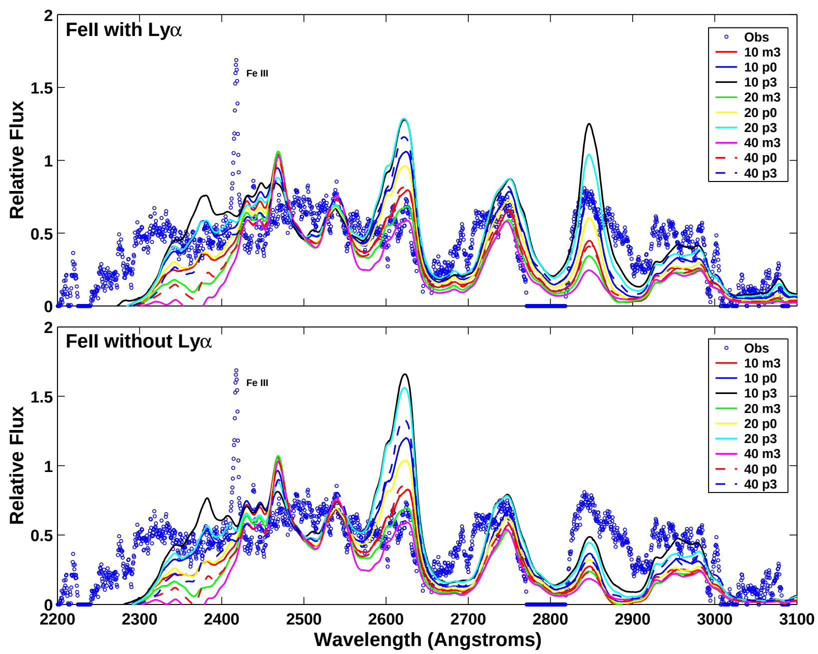
R-MATRIX CODES: VARIOUS STAGES

- **R-matrix calculations can have 3 branches to proceed - 1) LS coupling & relativistic Breit-Pauli, 2) Large configuration interaction LS coupling, 3) Dirac relativistic**
- **Generates - Hamiltonian Matrix, Dipole Matrices, Bound Wave Functions, Continuum Wave Functions**
- **End results - 1) Energy Levels, 2) Oscillator Strengths, 3) Photoionization Cross sections, 4) Recombination Rate Coefficients, 5) Collision Strengths; - Astrophysical Models**

THE R−MATRIX CODES AT OSU



Results: Observation & Modeling: Emission spectra of Fe I - III in active galaxy 1 Zwicky 1. (Sigut, Pradhan, Nahar 2004)



- Blue - Observation; Curves - Various Models with 1000 energy levels, millions of transitions
- With (top) and without (bottom) Lyman-alpha fluorescent excitation of Fe II by recombining H-atoms. The models reproduce many of the observed features, but discrepancies indicate need for more accurate calculations.

NANOSCIENCE

X-ray Spectroscopy for Biomedical Application - Cancer Theranostics Research (2004-)

(Support: Large Interdisciplinary Grant from OSU)

NOTABLE FACTS:

CANCER RISK:

• Men - Lifeitme probability of developing cancer, by site during 2000-2002

Sites	Risk
All sites	1 in 2

Cancer (risk in order): Prostate, Lung and rectum, Urinary bladder, Non-Hodgkin lymphoma, Melanoma, Kidney, Leukenia, Oral Cavity, Stomach

• Women, US - Lifeitme probability of developing cancer, by site during 2000-2002

Sites	Risk
All sites	1 in 3

Cancer (risk in order): Breast, Lung and branchus, Colon and rectum, Uterine corpus, Non-Hodgkin lymphoma, Ovary, Melanoma, Pancreas, Urinary bladder, Uterine cervix

BROADBAND RADIATION IMAGING:

• includes X-ray, CT, PET, and other nuclear imaging modalities
• Used in screening, diagnostic work-up, image-guided

biopsy and therapy delivery

- X-ray and CT: broadband radiation, 20-140 keV, typically 80- 120 keV (Depends on changes in tissue density to detect soft tissue abnormality)
- Photoionization in bones, Compton scattering in tissues

ENERGY RANGE SELECTION:

- Compromise between Image Contrast and Patient Dose (absorption)
- Lower energy - Greater contrast in transmission radiograph - but insufficient penetration by absorption of intervening tissues
- Higher exposure and doses are needed due to linear absorption

RADIATION THERAPY:

- Mostly delivered by linear accelerators at 6-25 MeV (broadband with spectral peak at $\sim$1/3 of maximum accelerator energy)

Existing Radiation and Chemical therapies are inefficient and largely ineffective
(e.g. Nature Reviews: Cancer, Vol. 5, March 2005)

OUR AIM: A PARADIGM CHANGE

- NanoSpectroscopy with NanoTechnology
- X-ray Broadband to X-ray Spectroscopy

Absorption and emission of X-rays are highly efficient at narrow resonant spectral energies

3-Step Process of Treatment

(1) Narrow-band (pulsed) X-ray impact on high-Z nanoparticles embedded in malignant tissues

- Nanoparticles - heavy elements, not abundant in living tissues (e.g. C, O, Fe, etc), non-toxic after injection, and tumor-seeking: Br, I, Gd, Pt, Au
- Higher energies ($\sim$50 keV or higher) deliver less harmful dose to normal tissue in front of the target heavy metal

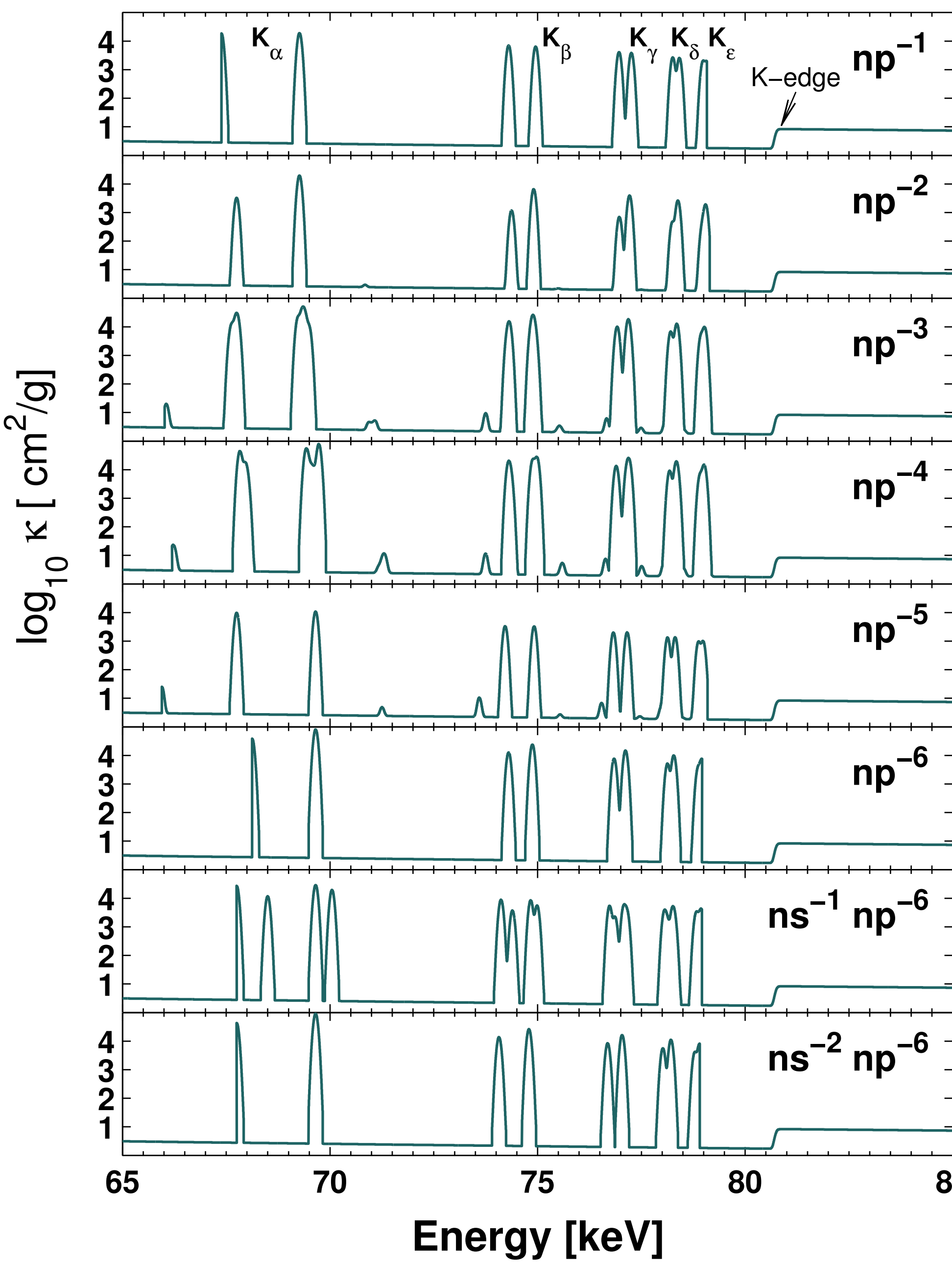
(2) Ionization - (in situ) X-ray radiation + Electrons, recombination $\rightarrow$ X-ray radiation

(3) Auger Cascades $\rightarrow$ photons and electrons and in vivo destruction of malignant DNA cells

- Interface Atomic & Molecular Spectroscopy, Biomedical science

X-ray Spectroscopy in Cancer Research:
X-ray Mass Absorption (κ) by Nano Gold Particles with 2s, 2p-Subshell Vacancies: 1s-np K-Shell transitions

The K-complexes of resonances, in E = 67.5 - 79 keV, show photo-absorption exceeding the background below the K-edge ionization by large factors (Pradhan et al 2008)



Cancer Treatment with Gold X-rays

(Hainfeld, Slatkin, Smilowitz 2004)

N312 J F Hainfeld *et al*

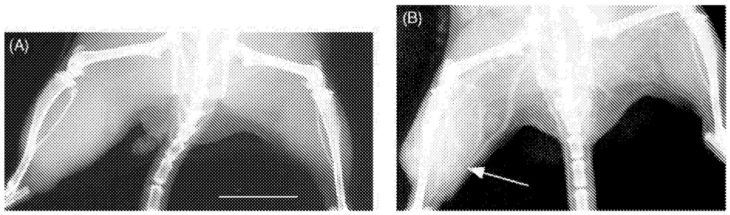


Figure 3. Radiographs of mouse hind legs before and after gold nanoparticle injection. (A) Before injection. (B) 2 min after i.v. gold injection (2.7 g Au/kg). Significant contrast (white) from

Gold nanoparticles for radiotherapy in mice N311

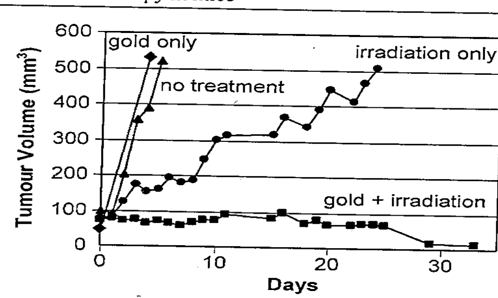
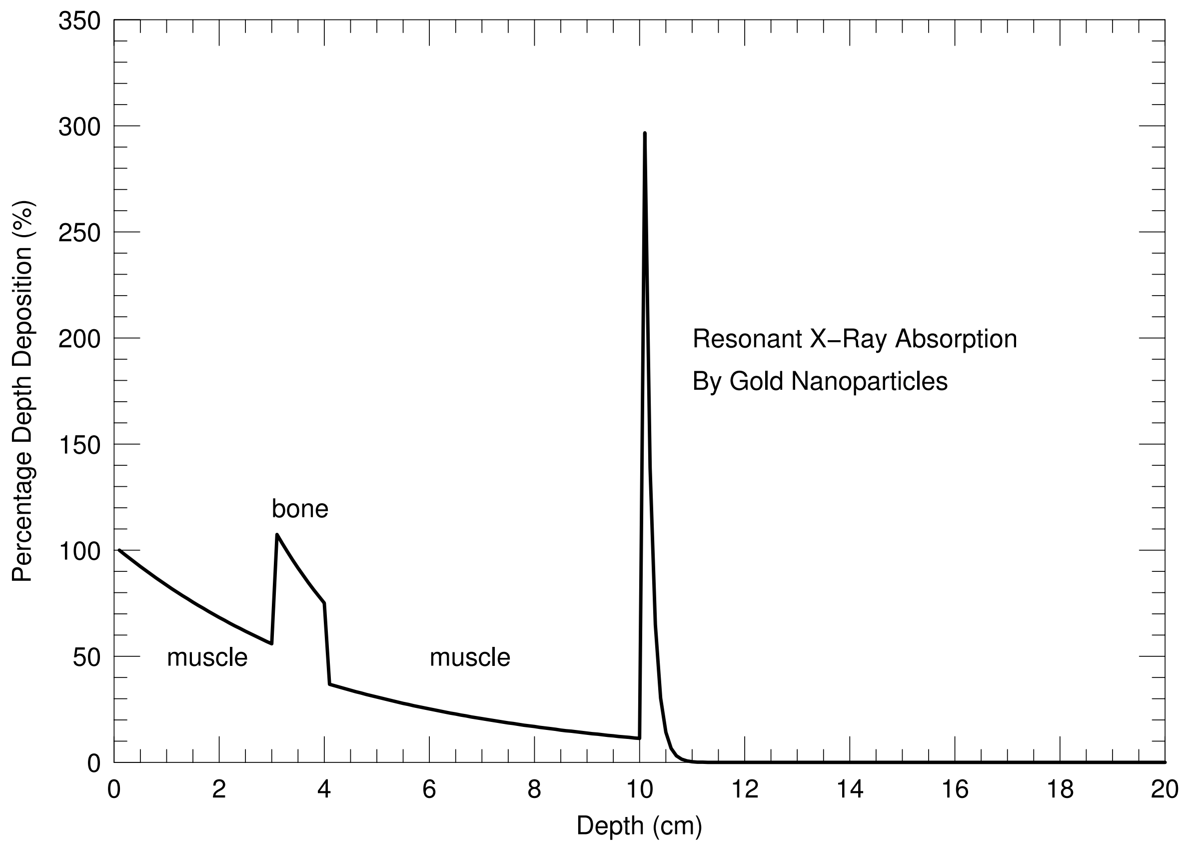


Figure 1. Average tumour volume after: (a) no treatment (triangles, $n = 12$); (b) gold only (diamonds, $n = 4$); (c) irradiation only (30 Gy, 250 kVp, circles, $n = 11$); (d) intravenous gold injection (1.35 g Au/kg) followed by irradiation (squares, $n = 10$).

- Top figures shows radiograph of mouse hind leg before and after injection of gold nanoparticles
- X-ray emission from doped gold nanoparticles in malignant cancer tissue is found to kill the defective cells with less radiation than used in radiation therapy
- 30 days experiment found that irradiation with gold nanoparticles controlled the tumor volume.

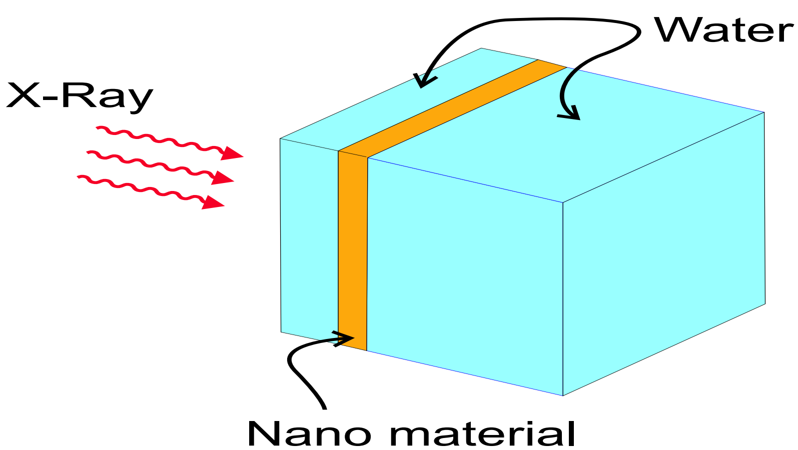
Simulation of Resonant K$_\alpha$ X-ray (68 keV) Absorption by Gold Nanoparticles in Tissues (Pradhan et al. 2008)

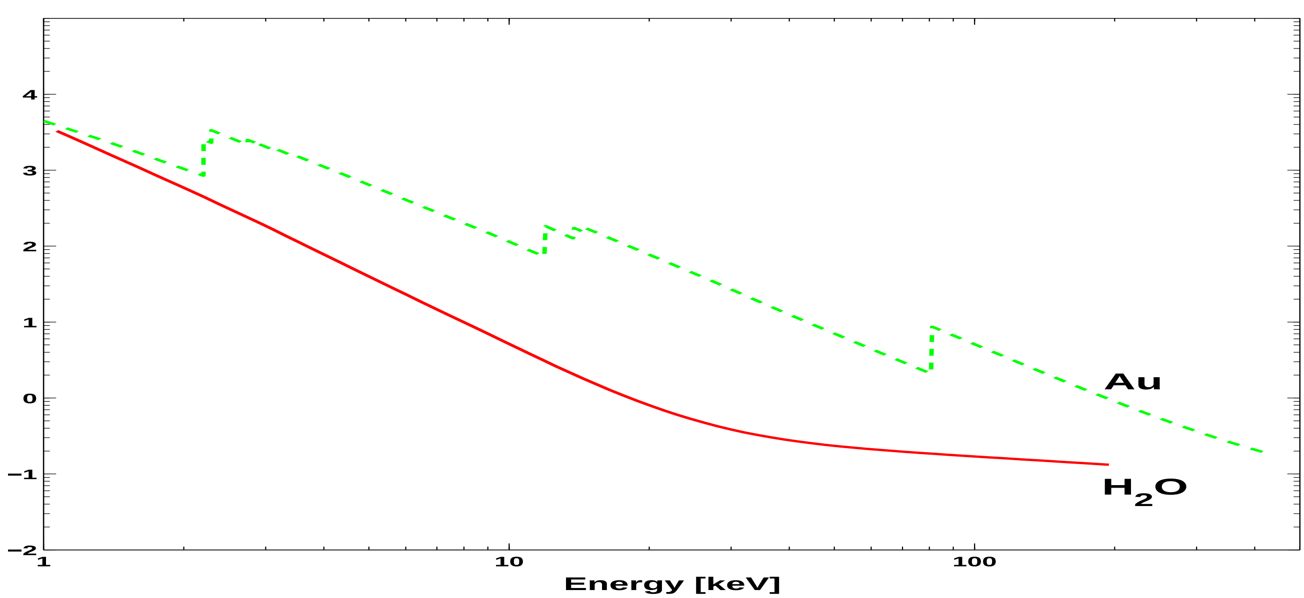


- Tumor Depth = 10 cm, Gold layer concentration = 0.001 cm/g
- Percentage Depth Deposition (relative to background) of 68 keV X-rays due to partial $K\alpha$ attenuation by gold nanoparticles embedded in body tissue at tumor site 10 cm inside the surface.
- Complete absorption of X-rays within < 1 cm of the Au-layer (the numerical simulation assumes uniform distribution).

GEANT4 SIMULATION (Monte Carlo): K-EDGE (80 keV) EFFECT ON X-RAY ABSORPTION BY GOLD NANOPARTICLES IN WATER (Pradhan et al. 2008)

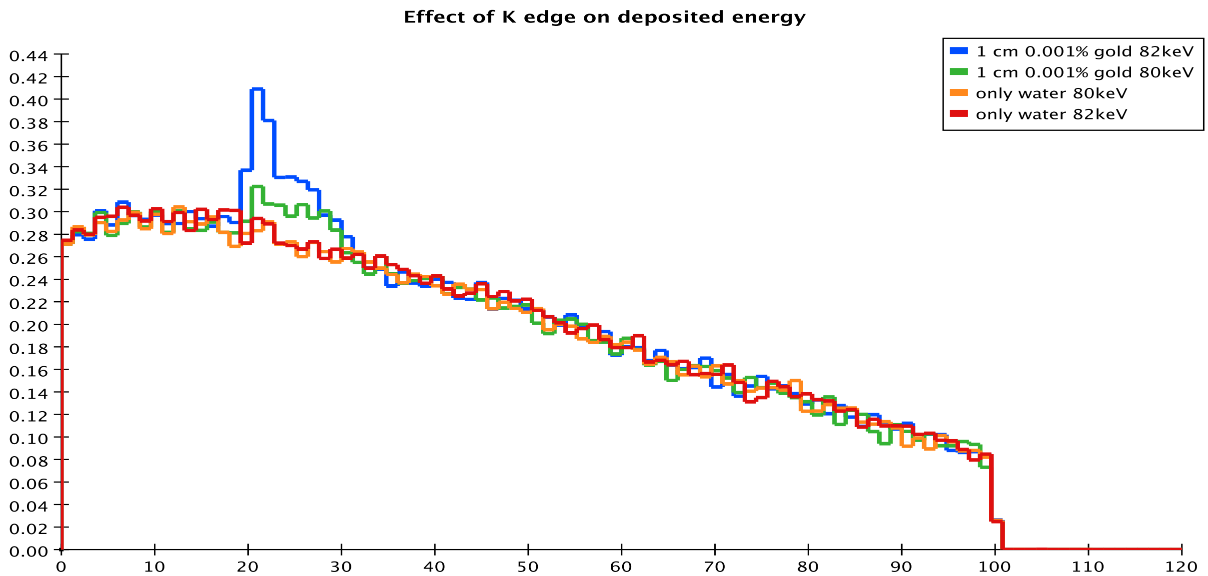
Figure (Top): - X-rays travelling in a water cube with a thin film (1 mm/g) of Gold nanoparticles
Figure (Bottom): - Photoinization cross sections of gold and water; Gold K-edge at 80 keV

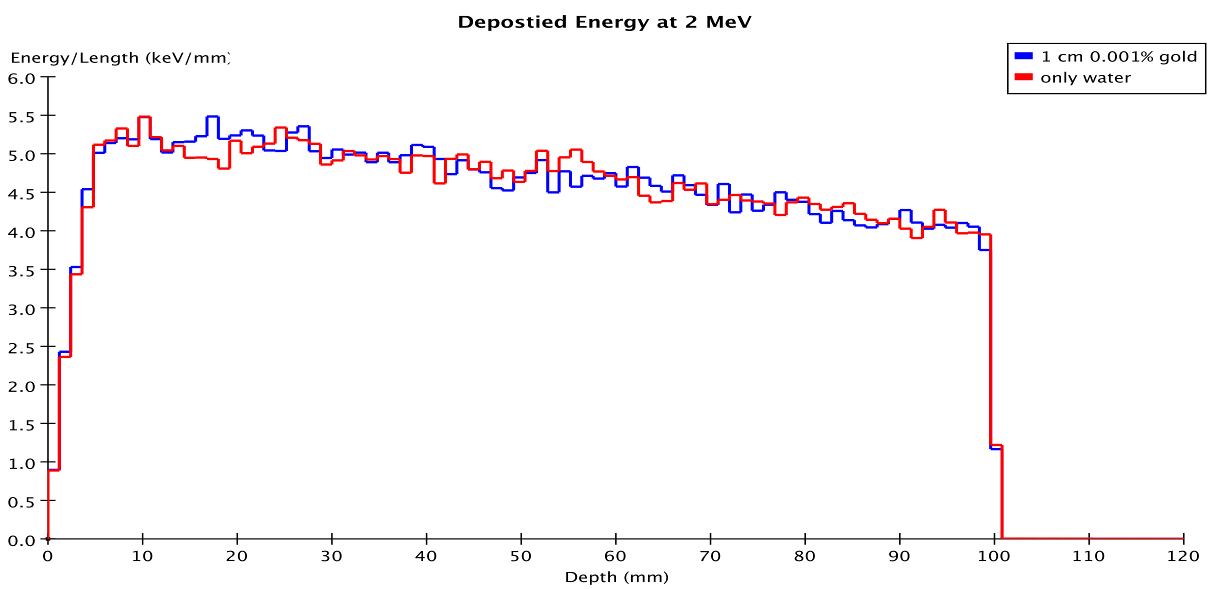




GEANT4 SIMULATION: X-RAY ABSORPTION BY Au

Figure (Top): - Enhanced (50%) absorption of X-rays (blue) at Au K-edge over H_2O (red)
Figure (Bottom): - 2 Mev high energy X-rays are transparent to Au (blue) and H_2O (red)





Research Presentations & Publications (in Nanotechnology, physics, and bio-medical science)

1. **"Resonant Enhancement in K-shell X-ray Absorption in High-Z Plasmas: Attenuation by Iron and Gold Ions", Anil K. Pradhan, Sultana N. Nahar, M. Montenegro, Y. Yu, Chiranjib Sur, M. Mrozik, R. Pitzer (submitted, 2008)**
2. **"Oscillator strengths and radiative transition rates for K_α lines in gold X-ray spectra: 1s-2p transitions", Sultana N. Nahar, Anil K. Pradhan, Chiranjib Sur, J. Quant. Spec. Rad. Transfer 109, 1951 (2008)**
3. **"Resonant X-Ray Attenuation by Highly Ionized Ions of High-Z Elements", Anil Pradhan, Sultana Nahar, Yan Yu, C. Cur, M. Montenegro, M. Mrozik, R. Pitzer, in the** *39th Annual Meeting of the APS Division of Atomic, Molecular, & Optical Physics (DAMOP)*, **May 27-31, 2008; State College, Pennsylvania, Bull. Am. Phys. Soc. B6.00001**
4. **"Resonant X-ray Irradiation of High-Z Nanoparticles For Cancer Theranostics" (refereed presentation), A Pradhan1, S Nahar2, M Montenegro3, C Sur4, M Mrozik5, R Pitzer6, E Silver7, Y Yu8 *, (1) Ohio State University, Columbus, OH, (2) Ohio State University, Columbus, OH, (3) Ohio State University, Columbus, OH, (4) Ohio State University, Columbus, OH, (5) Ohio State University, Columbus, OH, (6) Ohio State University , Columbus, OH, (7) Harvard University, Cambridge, MA, (8) Thomas Jefferson University, Philadelphia, PA, SU-GG-J-212, 50th Annual Meeting of the American Association of Physicists in**

Medicine in Houston, TX from July 27 - 31, 2008 (Joint Imaging-Therapy General Poster Discussion)

5. **"Innovative Instrumentation for Resonant Cancer Theranostics E Silver1 *, A Pradhan2 , Y Yu3 , (1) Harvard University, Cambridge, MA, (2), Ohio State University, Columbus, OH, (3) Thomas Jefferson University, Philadelphia, PA, 50th Annual Meeting of the American Association of Physicists in Medicine in Houston, TX from July 27 - 31, 2008**

6. **"Resonant X-ray Irradiation of High-Z Nanoparticles For Cancer Theranostics", A.Pradhan, S. Nahar, M. Montenegro, C. Sur, M. Mrozik, R. Pitzer, Y. Yu, E. Silver, 3rd Annual** *Ohio Nanotechnology Summit*, **April 24-25, 2007, Akron, Ohio, Poster Sessions and Abstracts, NB-3, p.37**

7. **"Resonant X-ray Irradiation of High-Z Nanoparticles For Cancer Theranostics", Anil Pradhan, Sultana Nahar, Max Montenegro, Chiranjib Sur, Mike Mrozik, Russ Pitzer, Yan Yu, Eric Silver,** *Ohio: The Global Pioneer in Biomedical Imaging*, **October 19, 2007, Ohio State University, Columbus, Ohio; Poster Presentation**

8. **"Nanospectroscopy of Materials and biomedicine at fundamental atomic and molecular scales", M. Mrozik, R. Pitzer, J. Oelgoetz, M. Montenegro, A.K. Pradhan, B. Larkins, 2nd Annaul Ohio Nanotechnolgy Summint, Columbus, April 4-5, 2006**

9. **"Nanospectroscopy of Materials and biomedicine at fundamental atomic and molecular scales", A.K. Pradhan, S.N.**

Nahar, R. Pitzer, P. Sadayappan, J. Oelgoetz, R. Tyagi, B. Larkins, W. Eissner, Y. Yu, M. Schell, 1st Annual Ohio Nanotechnolgy Summint, Dayton, Ohio, March 2-3, 2005

CONCLUSION

1. **Spectroscopy holds the key to understanding of astronomical objects: It provides the diagnostics of various physical and chemical conditions of the astrophysical plasmas**

2. **X-ray spectroscopy can be directed to non-invasive treatment of cancer.**

3. **X-ray absortion and emission of gold nanoparticles have shown effective destruction of malignant cells in mice.**

4. **Knowledge of spectroscopic resonant energy positions and enhancement are crucial to study of X-ray absorption.**